Multiscale Density Functional Theory and Computations

Enabling system-level simulations of nanoscale systems

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Summary

Density functional representation is the basic modeling tool of nanoscience. Solving the optimization problem associated with this representation is the key to predicting the chemical and mechanical properties of nanoscale systems. We use multiscale mathematics to expand the reach of computational approaches from the thousands-of-atoms configurations that they can currently solve to the millions-of-atoms configurations that they need to solve.

Nanoscience, the science related to the understanding and control of the matter at the nanoscale, the length scale of 1–100 nm, carries the promise of significantly improving our everyday life, in the immediate and medium-term future. Using nanosciene to obtain novel nanocatalysts, for example, will result in far-improved catalytic converters and substantially reduced pollution from our cars. The all-important aim of clean energy depends heavily on the ability of efficiently storing hydrogen storage, for which nanoscience is expected to provide a broad range of solutions.

Nanoscience allows access to a vastly enhanced array of new material properties. The electronic structure of materials undergoes dramatic changes when their dimensions are reduced to the nanoscale regime, leading to new regimes of physical, mechanical, and chemical behavior not observed in bulk materials. For example, nanoscale oxide structures exhibit unusual characteristics compared with their bulk counterparts: thermodynamic stability and lattice properties, magnetic properties, ion transport, optical properties, chemical reactivity, and ferroelectric properties. Nanoscale metals, polymers, and semiconductor materials have also been shown to exhibit unique properties and a wide range of technological applications .

A key endeavor in unlocking the potential of nanoscience is the ability to use high-performance computing to predict the behavior of nanoscale materials to a high accuracy. When the chemical properties of such materials are sought, as is the case in virtually all energy-related applications, the distribution of electrons inside of the material must also be correctly determined. Therefore, any computation must accurately solve the density functional theory (DFT) problem whose result is the distribution of electrons (the electron density) at the surface and inside the nanoscale material. For large nanoscale systems (that may have millions of atoms), such computations are outside the reach of stat-of-the-art computational

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techniques for DFT. Such techniques currently take several weeks on 64-processor machines for configurations with a few thousand atoms.

To remove this bottleneck, we developed a multiscale approach that computes the electron density in the nanoscale system, while carrying out detailed computations only in small subdomains, each of which is of a size that is approachable by current DFT methods. The aggregate number of atoms in the subdomains is only a fraction of the total number of atoms, which makes the overall computation within reach in the immediate future. In Figure 1, we present a comparison between the multiscale approach and the direct simulation approach for a one-dimensional, Thomas-Fermi DFT problem. We see that the multiscale approach (interpolation reconstruction) produces virtually indistinguishable results while using less than half the number of degrees of freedom (mesh points) to carry out the computations. The results validate our rigorous mathematical analysis of the approach.

We are currently investigating extensions of our technique to generic DFT approaches, such as the highly accurate Kohn-Sham approach. In addition, we are pursuing advanced techniques for efficiently solving the resulting very large scale optimization problem. To harness the potential of our advances, we are building the three-dimensional multiscale DFT code MS-DFT. Computations of the forces on the nuclei in a 1000-atom subdomain of the nanoscale material is presented in Figure 2. We expect that the code will allow us to accurately compute millions-of-atoms configurations while effectively carrying out computations in domains that have an aggregate of tens of thousands of atoms. This would put nanoscale system prediction and design within reach in the immediate future. The approach is developed in collaboration with scientists from the Materials Science Division at Argonne.

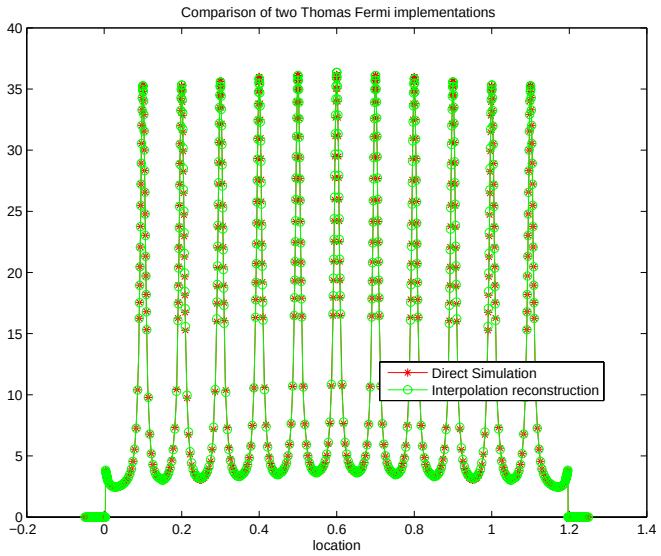

Figure 1: Comparison of the multiscale technique with the direct numerical simulation technique; electronic density plot

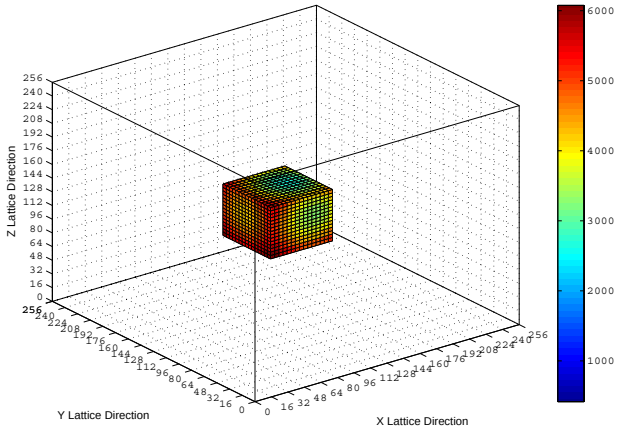

Figure 2: Computation of the forces on the nuclei with the MS-DFT code

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